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TRENDS IN INSTRUMENTATION FOR ENVIRONMENTAL RADIATION MEASUREMENTS AT LOS ALAMOS SCIENTIFIC LABORATORY

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Recent instruments developed to fulfill radiation monitoring needs at Los Alamos Scientific Laboratory are described. Laboratory instruments that measure tritium gas effluents alone, or in the presence of activated air from D-T fusion reactors are discussed. Fully portable systems for gamma, x-ray, and alpha analyses in the field are described. Also included are descriptions of survey instruments that measure low levels of transuranic contaminants and that measure pulsed-neutron dose rates.

Introduction

A wide assortment of radiation monitoring instruments has been developed at Los Alamos Scientific Laboratory (LASL) in recent years. These developments have been confined to those needs that cannot be filled by commercially available instruments. This paper reviews some of our instruments that have applications in the broad classification of environmental measurements and discusses current problems involving instruments.

The descriptions are grouped in three major categories. The first pertains to instruments and problems relating to the monitoring of airborne tritium. Tritium is widely used in research programs at LASL, and we have a continuing instrumentation program to control, minimize, and document atmospheric releases of this isotope. The second category includes portable instruments used in field survey applications. These instruments have been designed with high sensitivity as a key feature and with physical characteristics desired by the people who do the field work. The third category includes instruments for nuclear spectroscopy with emphasis on portability and sensitivity.

Airborne Tritium Monitors

A tritium beta has a maximum energy of 18 keV with a mean energy of 5.6 keV. At these low energies, the sampled air must be in intimate contact with the detecting medium. Most of our instruments use flow-through ionization chambers, although we are developing a flow-through scintillation detector that is described below. The simplest measurement is to determine total tritium activity without regard for its form (usually HT gas or tritiated water vapor), or whether it is mixed with contaminant isotopes. We describe several instruments in this nonselective class, along with preliminary designs for an instrument that differentiates between tritium gas and tritiated water vapor (HTO), and one that selectively measures tritium in the presence of gases emitting higher energy betas in the activated air produced by D-T fusion reactors.

Nonselective Tritium Monitors

Instruments that detect total tritium regardless of its form are common because they are relatively simple to fabricate, and measurements of total tritium will usually provide information sufficient for radiological control. The readings in dosimetric terms are usually in the "safe" direction because exposure tolerances assume worst-case form (HTO) in the sampled air. Note that the radiological significance of these forms varies greatly: exposure to a given concentration of tritium gas (HT) in air leads to a dose that is hundreds to tens of thousands of times less than that resulting from exposure to the same concentration of HTO.

We have developed special ionization chambers and electronics for our monitoring needs. The chambers are to provide reduced sensitivity to contamination and extended radiation sensitivity range. Important features of the electronics are stability and wide range.

A popular flow-through tritium detector is the Kanne chamber. Figure 1 shows a conventional 51-ℓ

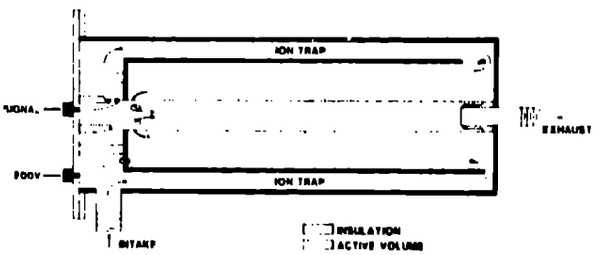

Fig. 1. Conventional 51-ℓ Kanne chamber.

Kanne chamber, as described by Hoy,[1] which consists of three concentric cylinders, with the outer and inner (signal electrode) cylinders at or near ground potential and the intermediate cylinder at about 200 V. The inner region between the signal electrode and the intermediate cylinder is the measuring chamber. However, in our operating environments, the Kanne chamber is exposed to high concentrations of radioactive gases such as HTO, or to air contaminated with tritiated oil, and the build-up of surface activity reduces the sensitivity of the chamber at low tritium concentrations. Many of our environmental measurements require integration of the detector current to determine the total tritium effluence with time; any uncertainty in detector background or instrumental zero degrades these measurements.

To cope with these problems, we redesigned the chamber and developed new electronics.[2] Figure 2 shows the improved Kanne chamber. Our objective was to reduce the sensitive surface area, where contamination contributed to chamber background. The intermediate high-voltage cylinder was replaced by a cylindrical array of 45 nichrome wires that are parallel to the center electrode. The conventional central collecting electrode was replaced by a rod 1/10 the original diameter. All surfaces outside the wire cylinder are greater than the maximum tritium beta range

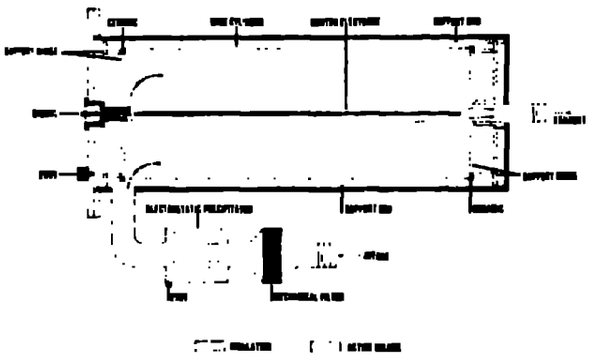

Fig. 2. Improved Kanne chamber.

(about 1 cm) from the wires. The chamber's area sensitive to contamination was reduced by a factor of 40. Because this design eliminated the internal ion trap, an external deionizer is provided.

The new electrometer-chargemeter, shown in Fig. 3, was designed to measure currents as low as 1 fA (10^{-15} A), and to integrate these currents to measure accumulated charge. The electrometer has 4 decades of range, with a switch to select current ranges. Logarithmic and linear display of current is furnished in analog format. The linear signal from the electrometer feeds a low-drift voltage-to-frequency converter to provide digital signal integration for charge measurements. Chargemeter readout is a digital display that covers 10 decades from 10^{-12} C/digit to 10^{-3} C full scale. Readout is with 3 decades of digital indicators and exponent multipliers. The varactor bridge electrometer operational amplifier and its associated high-megohm resistors are housed in a separate temperature-controlled oven, shown on the left in Fig. 3. This is a standard component oven modified to stabilize at a temperature just above maximum expected ambient temperature. In this way, we achieve the necessary instrument sensitivity and stability for accurate long-term effluent integration. The oven assembly is placed as close as possible to the ionization chamber signal connector to minimize spurious cable effects. The control and readout chassis may be as far as 50 to 100 m from the point of measurement. This feature is particularly useful for stack and duct monitors.

One of these systems was tested at one of our tritium processing laboratories that is noted for its contaminating environment. After 16 weeks of constant use, no detectable change in the measurement baseline was observed, while considerable interior contamination was observed in a standard Kanne chamber in series with the new chamber. Steady-state tritium concentrations of about 0.1 $\mu Ci/m^3$ would be measurable, even with the contaminating environment. Note that the present derived air concentration (DAC)* for controlled areas for HTO is 5 $\mu Ci/m^3$.

In many cases, small concentrations of total tritium must be monitored routinely, but the system must be able to react to sudden, large excursions such as in an accident. Therefore, we are developing a monitor system that is sensitive to low concentrations of tritium and, at the same time, is linear over a dynamic range of 10^8. The detector will have the same resistance to contamination as that in the improved Kanne chamber.

Figure 4 shows a cross section of the wide-range ionization chamber we are building. The measuring region uses parallel grid electrodes that provide improved collection fields over those of cylindrical chambers. The grids are made of open wire mesh to reduce the surface area that can be contaminated. Each of the two active regions sends its own collecting electrode signal to a separate electrometer. The first region has an active volume of 1 ℓ and is used on the low range (approximately 1 to 10^4 $\mu Ci/m^3$). The volume of the second region is 0.1 ℓ and covers the high range (10^4 to 10^8 $\mu Ci/m^3$). The high-range section has closer grid spacing to reduce recombination at high concentrations. The incoming air can be preheated to reduce contamination and hysteresis.

Figure 5 is the block diagram for the current and charge-measuring electronics to be used with the wide-range detector. The two electrometer amplifiers receive current signals from the appropriate ion-chamber volumes. The low-range electrometer will be similar to that described above. The temperature-controlled enclosure will assure long-term zero stability to allow digital integration of low tritium concentrations

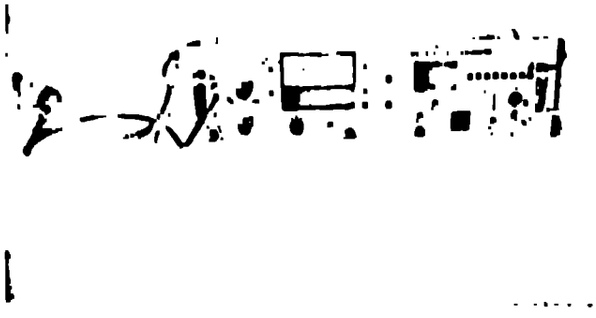

Fig. 3. Electrometer-chargemeter used for low-drift measurements with flow-through ionization chamber.

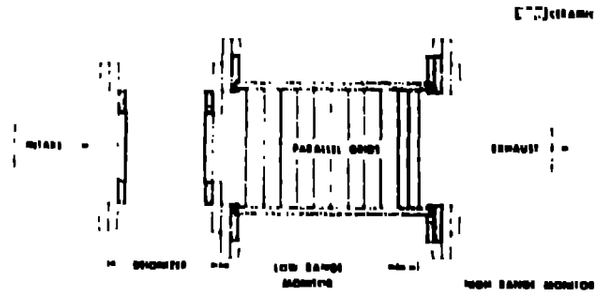

Fig. 4. Wide-range ionization chamber with two regions for signal collection. Each region has its own output connector (not shown).

*This term replaces maximum permissible concentration in air (MPCa).

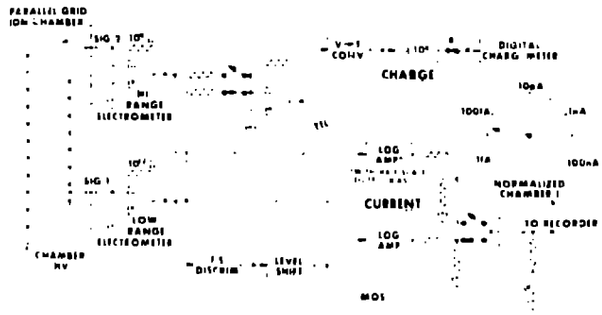

Fig. 5. Block diagram for processing current signals from two outputs from wide-range ionization chamber.

for accurate charge measurements. The high-range electrometer will be a low-cost FET type, with the transimpedance resistor scaled appropriately for the range desired and for the smaller chamber volume. Each amplifier feeds its own 4-decade logarithmic amplifier. A solid-state switching arrangement (all switches at low impedance levels to prevent transient excursions) connects only one input signal at a time to the current and charge-measuring circuits. The crossover sensing between the two sets of circuits is done with a discriminator that triggers at the full-scale level on the low-range electrometer. Suitable output analog biasing of the high-range current signal and proper scaling factor of the digital charge pulses will assure continuity of readings over the 8 decades.

HT/HTO Selective Monitor

Radiological effects of tritium vary greatly with its form. Therefore, to avoid gross overestimates of hazard due to exposure to tritium gas, each form of airborne tritium should be measured to obtain a more accurate measure of the radiological hazard. The goal, then, is to measure a small amount of the most hazardous substance (HTO) in real time in the presence of a large quantity of HT. Semipermeable membranes have been used[3] to improve the HTO:HT ratio but the technique is rather insensitive, and measurement of small concentrations takes more than 10 min. Another technique is to measure the total tritium, remove the HTO with a desiccant, and measure again. However, if a relatively large amount of HT is present, one must determine a small number (the amount of HTO) by subtracting two large numbers.

We propose to measure the HTO by humidifying the air and then removing the H_2O and HTO by condensing it it onto a cooled scintillator, as shown in Fig. 6. This scintillator is a rotating disk that passes between two photomultiplier tubes (PMT) before being dried and cooled again. The process is continuous, with an expected response time of 15-30 μs. The condensed water insulates the scintillator from the HT gas, thus giving a direct measurement of the HTO alone. This system should be sensitive to concentrations of a few to 10^4 $\mu Ci/m^3$ of HTO.

Tritium/Contaminant-Gas Selective Monitor

Tritium is a radionuclide of primary concern in assessments of the potential exposures to workers and the general public due to fusion research and development.[4] The D-T neutrons from fusion reactors will produce high concentrations of radioactive gas contaminants (primarily ^{13}N, ^{16}N, and ^{41}Ar) in the air sur-

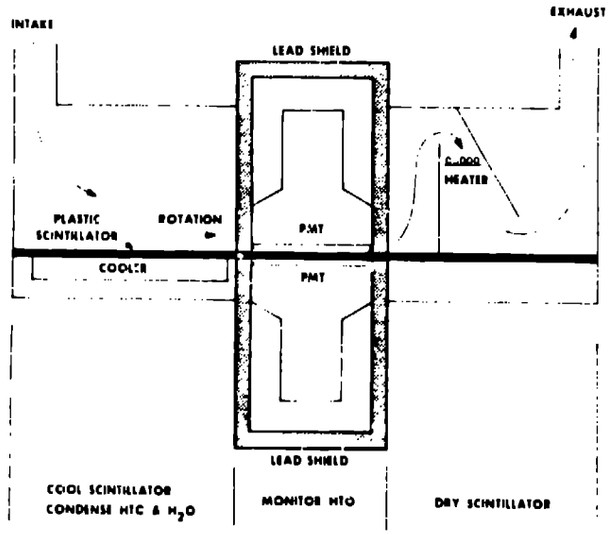

Fig. 6. Tritiated water-vapor monitor sketch. HTO condenses on the rotating sheet of plastic scintillator that brings the activity between the PMTs for counting.

rounding the reactors. Ordinary tritium air monitors such as those described above will be sensitive to this activated air, making detection of a tritium release following a burst of neutrons difficult or impossible. Therefore, it is imperative to have an instrument that can discriminate between tritium and the contaminant gases. A survey of existing and proposed methods of solving this problem[5] shows that commercially available proportional counter systems provide very good sensitivities and good discrimination if the contaminating gases are known. However, these detectors need a counting gas, and have a limited range of sensitivity at higher tritium concentrations.

We use a concentric flow-through ionization chamber system, which is relatively simple, can handle a wide range of concentrations, and costs less than commercial counting systems. The disadvantages include lower sensitivity and less discrimination than the proportional counter. Figure 7 shows the three principal coaxial elements. Sampled air is brought into the split hollow central electrode. A partition from end to end allows half the electrode to serve as an air injector to the sampling volume, while the other half is an air collector for the flow-through action. A series of holes along the length of the electrode gives uniform distribution of air injection and collection along the chamber axis. The intermediate cylinder around the central electrode acts as a partition between the inner measuring region and the outer compensating region bounded by the compensating chamber wall. The intermediate cylinder acts as a common signal electrode at ground potential.

Discrimination is accomplished as follows. The intermediate cylinder is fabricated to be air tight, with a wall of thin Mylar film stretched on a wire grid framework. This common wall between inner and outer chambers stops tritium betas but allows higher energy betas to penetrate to the outer chamber. Thus, the inner measuring chamber will have currents produced by tritium and energetic beta ionization, while the outer compensating chamber will have energetic beta currents only. The high voltage on the central electrode is of opposite polarity to that of the outer compensating chamber wall, so the current collected by the intermediate signal electrode will

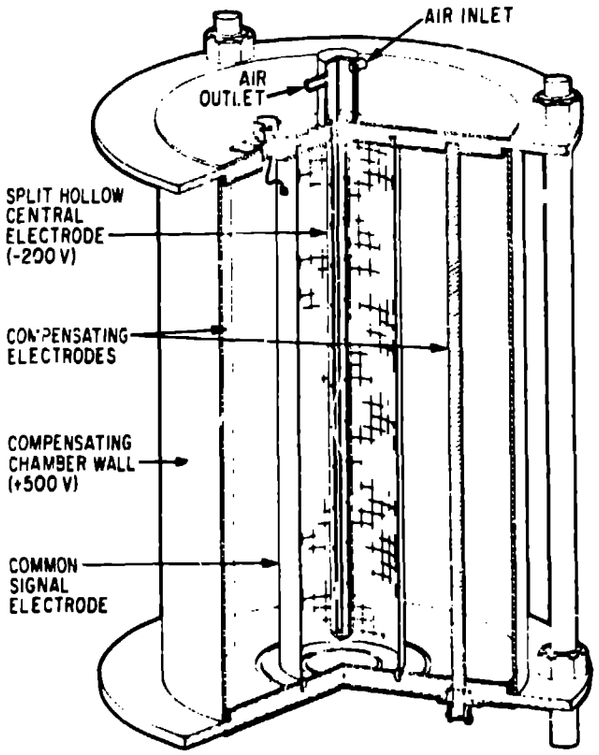

Fig. 7. Cross section of flow-through ionization chamber that selectively measures tritium in the presence of high-energy radioactive gases.

by the difference signal of the two chambers, which is a measure of tritium concentration. We carefully balanced the detector to reject the contaminating isotope by using volume-compensating electrodes in the outer region. The potential on these electrodes is adjusted to make the effective volume of the outer chamber equal to that of the inner chamber for the contaminating high-energy betas.

The theoretical limitations of this detector are established by statistical considerations and geometric end effects. For example, if 1000 $\mu Ci/m^3$ concentration of a high-energy beta gas were present in a 1-ℓ chamber feeding an electrometer with 30-s time constant, the minimum detectable concentration of tritium would be 5 $\mu Ci/m^3$. However, practical performance is limited by pressure and temperature variations and other noise, so that measured performance is about half the calculated performance. The mixed beta energies from activated air at fusion reactors is not a serious problem in balancing the chambers. The ^{13}N and ^{41}Ar contaminants have very similar beta energies, and ^{16}N has such a short half-life that it decays before our tritium measurements must be made. The effects of annihilation gamma radiation from ^{13}N positrons being absorbed at the compensating chamber wall remain to be studied.

Portable Instruments for Field Survey

Industry has been a prolific source of instruments for most portable survey meter needs. Nevertheless, LASL work sometimes creates a need for special instruments.

Transuranic Contaminant Survey Instruments

Because of LASL's heavy metals research and development, measurement of such environmental contaminants as plutonium and americium is important. For example, a special instrument was required when LASL was requested to conduct an environmental radiological evaluation on 5500 acres of land near LASL technical areas.[6] Because the property was to be opened to the public, extreme care was required in the study. The need was for a portable system to provide rapid field measurement for determining environmental levels of mixed transuranic contaminants, and the Los Alamos Field Pulse Height Analyzer (LAFPHA)[7] was quickly developed.

The LAFPHA used a commercial scintillation assembly consisting of a thin NaI (Tl) crystal on a cylindrical light pipe viewed by a 5-in. PMT. A beryllium entrance window allowed detection of x rays and low-energy gamma rays. The electronics consisted of six individually settable channels with appropriate counters, timers, and displays. One channel was set to span the energies at ~17 keV to detect L-series x rays of uranium occurring with the alpha decay of plutonium, and one channel was adjusted to detect the 59.5-keV gamma-ray emission of ^{241}Am. The other energy channels were set to give background correction readings. Figure 8 shows the instrument in use in the field. It detected surface activity less than 100 nCi/m^2 for ^{239}Pu and 20 nCi/m^2 for ^{241}Am.

Fig. 8. Los Alamos Field Pulse Height Analyzer.

To further improve our capability to measure low-level transuranic pollution, we developed a portable instrument[9] using a phoswich detector to decrease sensitivity to background and increase detectability of low-energy events. The phoswich scintillation detector (commercially available) comprises two different scintillators attached together and to a single PMT. The two crystals have sufficiently different phosphor decay times to allow determination by pulse-shape analysis of the spatial origin of the photon interaction. Hence, it can differentiate between desired low-energy events that stop only in the front

thin crystal, and those background events that deposit energy in both crystals.

Battery-powered electronics to analyze the signals from the scintillation detector were developed and several models of the survey instrument evolved; Fig. 9 shows the second model. The detector with its collimator is in a separate unit resting on the surface to be monitored. Instrument readout and controls are on the operator's chest. The backpack contains the batteries and most of the electronics. This instrument yields a reduction in background of about a factor of 3 compared to that of the LAFPHA, with little loss of sensitivity.

Alpha Survey Instrument

We developed a new portable alpha survey instrument, called the Wee Peewee,[9] that is smaller than comparable instruments and exhibits improved sensitivity. Figure 10 shows the instrument with the electronics package integral to the alpha air proportional probe. The commercially available probe is attached to the electronics case by two flanges and a spring clip to allow quick and easy change of probes in the field. The meter detaches from the case and can be extended by a coiled cord when the probe is placed so that the normal meter position is difficult to see. Most of the circuitry is fabricated in hybrid form, which contributes to the small package size. This technology has been transferred to industry, and a version of this instrument is now commercially available.

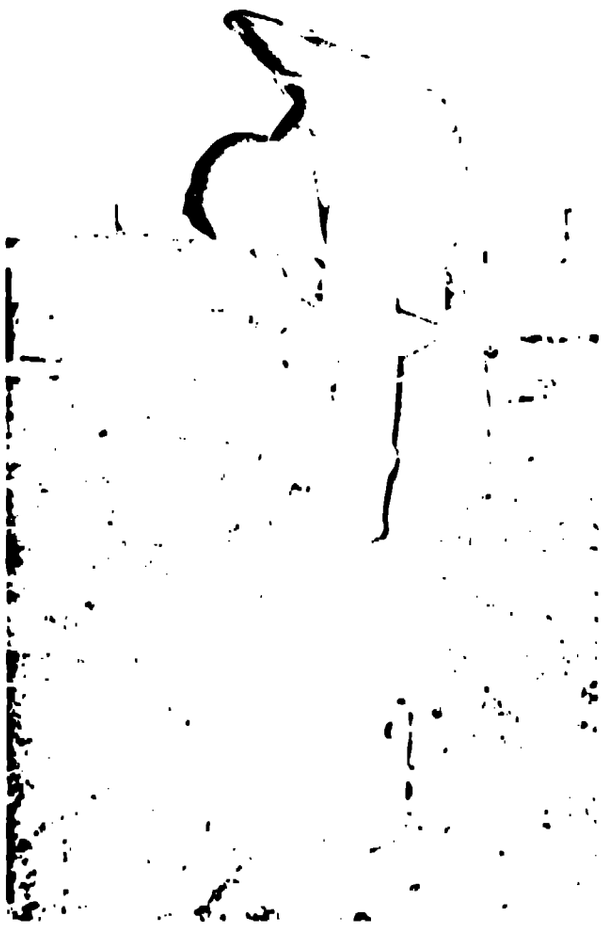

Fig. 9. Phoswich detector system in field use. Readout and controls are worn at the operator's chest. The backpack carries batteries and processing electronics.

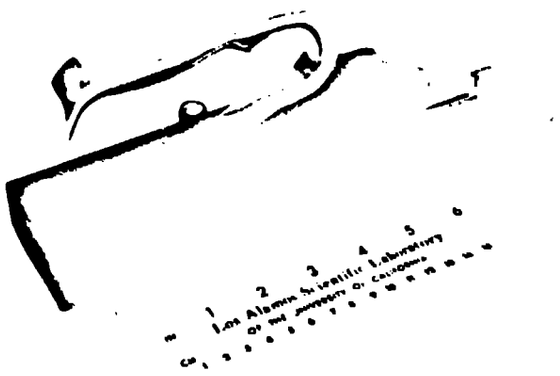

Fig. 10. Top view of Wee Peewee alpha survey meter with integral air proportional probe.

Pulsed-Neutron Dosimeter

Commercially available survey instruments cannot measure accurate pulsed-neutron dose rates in cases where the detector resolving time results in significant count losses. Although this is not an environmental monitoring problem that relates to exposures to a general population, it is significant because of the growing number of pulsed-neutron research facilities and the resultant potential occupational hazards.

About 10 years ago, workers at Fermi National Accelerator Laboratory developed a portable instrument, the Albatross III, that uses silver foil activation to measure dose-equivalent rates from pulsed neutrons.[10] The silver acts as an integrator of pulsed-neutron flux information, with the beta-activation product decaying at a rate that can be counted with a Geiger-Müller (G-M) tube detector. The activation half-life is short enough to follow changes in the neutron flux within several minutes. Albatross III had all the basic detector configuration and operational features we desired, but the calibration procedure of its analog circuitry was complex and lengthy. Therefore, we developed Albatross IV[11] with digital processing in a one-chip microcomputer replacing the analog weighting and averaging circuitry. We also added features to make the instrument more versatile and easier to calibrate.

Figure 11 shows the front view of the instrument. The G-M counters and foils are in the center of the polyethylene pseudosphere moderator. Readout on the log scale meter is from 1 to 300 mrem/h. Color-coded regions on the scale correspond to the colored status lamps on the top cover. The audio generator chirps with the detected neutron rate for normal conditions and the tone becomes continuous when the reading exceeds the alarm level set on the meter face.

Digital processing uses an Intel 8748 8-bit microcomputer. Its data and program memory are limited, but with careful programming they are sufficient for this application, and the result is a very compact and relatively inexpensive design. The

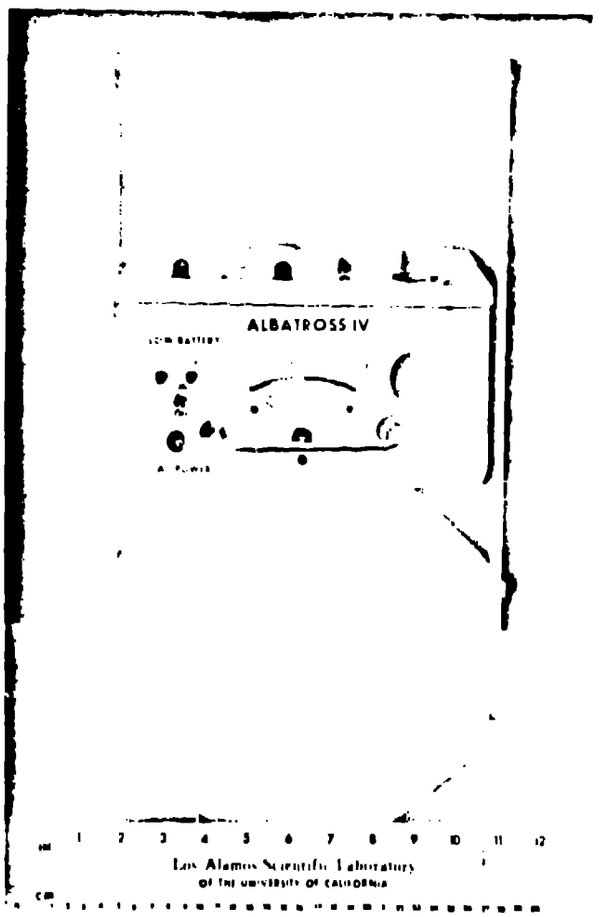

Fig. 11. Front view of Albatross IV pulsed-neutron survey instrument.

digital rate meter uses a running-average scheme that is easy to implement in the microcomputer. The overall data accumulation period (averaging time) is divided into 16 shorter time segments, and the count accumulated in each segment is stored in a rotating memory stack. The most recent count data replace the oldest at the end of each counting period. At this time, the total counts accumulated in the memory stack are updated. An averaging-time-multiplier control on the top cover is analogous to the time-constant control in an analog rate meter. The computer normalizes the net neutron count for each counting period by dividing it by the averaging-time-multiplier factor. Log conversion is done in the microcomputer, with a special digital-to-analog converter design driving the display meter.

Many Albatross IVs are being used to monitor neutron dose-equivalent rates in various experimental areas at the LASL Weapons Neutron Research Facility. This facility uses beams from 1 to 120 pulses/s with pulse widths of 1 ns to 500 μs. The 1-s averaging time permits observation of the radiation level in real time during beam tuning, and the longer averaging times are used to obtain precise readings.

Instruments for Nuclear Spectroscopy

We have developed fully portable field instruments for nuclear spectroscopy and a laboratory system with very high sensitivity for measuring transuranic contaminants in soil samples.

Portable Multichannel Analyzers

The need for completely portable spectrometers for environmental measurements is growing, and we have developed several models. The first system[12] is shown in Fig. 12. The multichannel analyzer (MCA) on the left is a full-capability pulse-height analyzer with 1024 channels, CRT display, and multiple region-of-interest integration. The instrument is microprocessor-controlled (Motorola 6802) with keyboard entry. Data are stored on an ordinary audio cassette recorder that is carried in the accessory case shown at the right in Fig. 12. The MCA signal input requirements allow many types of detectors to be used. Figure 12 shows a portable hyperpure germanium detector for high-resolution gamma spectroscopy and a standard NaI(Tl) scintillation detector that is normally carried in the accessory case. This system proved that an instrument with near-laboratory quality can be made completely portable.

An improved version of the first portable MCA, shown in Fig. 13, has all the basic features of the first model and additional operational conveniences. The accessory case is no longer needed; the cassette recorder is now integral with the MCA. Also, the high-voltage power supply and spectroscopy amplifier, now in the MCA case, are designed to be programmable. With the read-only memory (ROM) card that is plugged into the upper right corner of the instrument panel, the analyzer can be preprogrammed to perform certain tasks. This feature extends use of the instrument to untrained operators. The RS232 interface port was added for serial input/output communication with a separate computer or terminal. A version of this instrument will soon be commercially available.

Also in this category is the hand-held gamma-ray spectrometer gun,[13] shown in Fig. 14. The 2-kg package contains a NaI(Tl) scintillation detector, preamplifier, amplifier, high-voltage supply, and a 128-channel pulse-height analyzer. The system is microprocessor-based, and uses a single liquid-crystal display to indicate channel numbers and contents, region-of-interest integrals, and count rate in a region of interest. A separate display is used to show the MCA spectrum in segments.

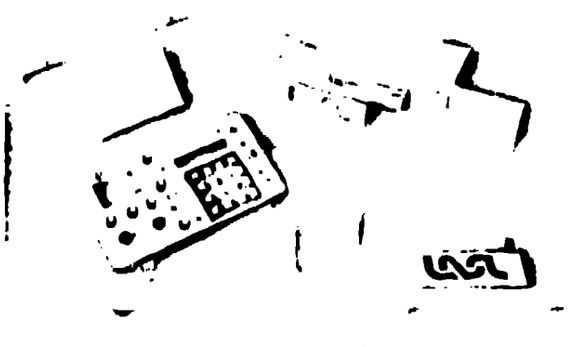

Fig. 12. The portable MCA is on the left; the portable HpGe detector is behind the standing NaI detector; the accessory case for the NaI detectors and the audio cassette recorder is front right.

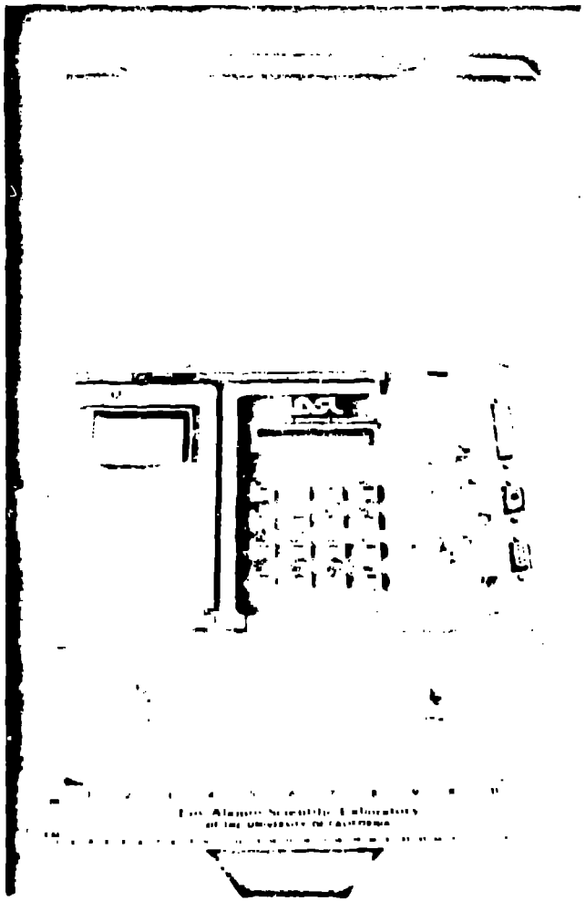

Fig. 13. Portable MCA, with scope display at top left, and liquid-crystal display at top center above key pad. The digital cassette recorder is to the left of key pad.

Low-Level Soil Radioassay System

We mentioned earlier the development of in situ environmental measurements on soil. For very low activity concentrations, soil samples must be brought to the laboratory for careful radioassay. Our new automated system[14] uses two commercial hyperpure germanium detectors to examine gamma-ray and x-ray emissions in the 0- to 200-keV energy range. The two detectors are mounted in opposition on either side of a thin sample of the soil to be assayed. Because plutonium is of particular interest, the L-series x-ray group from plutonium alpha decay is measured. Correction for any ^{241}Am that might be present is made. The pulse-height analyzer and the automated soil-sample changer are controlled by the same microcomputer that performs all calculations. Detection limits of approximately 15 pCi/g of Pu and 0.1 pCi/g of ^{241}Am have been demonstrated.

Acknowledgments

Many people at LASL contributed to the instrument developments and concepts described, and our thanks is extended to them all. Special thanks are given to C. J. Umbarger for his advice and liaison in these diverse activities.

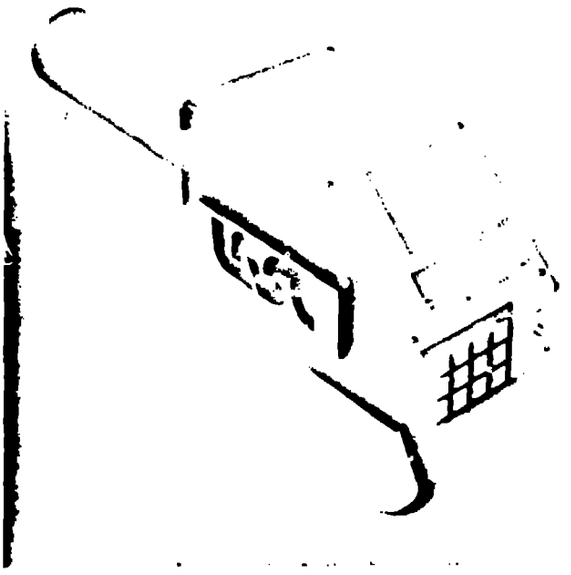

Fig. 14. Hand-held gamma-ray spectrometer. The scintillation detector extends to the left. The liquid-crystal assembly in the square opening displays the MCA spectrum in selected segments.

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